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Full length article

Polymer-specific dynamic probabilistic material flow analysis of seven polymers in Europe from 1950 to 2016

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ABSTRACT

Information on the flows of specific polymers through the anthroposphere forms the basis of any assessment of the environmental implications of plastic. This work presents a dynamic probabilistic material flow analysis of seven commodity thermoplastics: low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP), polystyrene (PS), expanded polystyrene (EPS), polyvinylchloride (PVC), and polyethylene terephthalate (PET). The analysis examines nine product sectors and 35 product categories, including textiles, across Europe from 1950 to 2016. Total consumption of these seven polymers was 90.0 ± 4.8 kg/cap in 2016. The in-use stock that year was 47 kg/cap for LDPE, 81 kg/cap for HDPE, 81 kg/cap for PP, 21 kg/cap for PS, 56 kg/cap for EPS, 163 kg/cap for PVC, and 16 kg/cap for PET. For EPS and PVC, the fraction contained in the in-use stock is 51% and 39% of the total production. Landfill is the major compartment where LDPE, HDPE, PP, PS, and PET end up (48%–60%). Elimination—incineration and destruction in waste incineration plants—occurs for 15%–19% of LDPE, HDPE, PP, PS, and PET, and 10% of EPS and PVC. Current European recycling rates vary from 11% for PS to 33% for PET. The precise separation of different polymers into very detailed product categories will allow the formulation of future scenarios about interventions to improve the circularity of specific polymers and provide the basis to assess the polymer-specific releases to the environment.

1. Introduction

Although plastics have brought many benefits, the plastics industry is now also seen as the source of many problems (Nielsen et al., 2020). There are many questions relating to plastic's environmental and human health impacts (Wright and Kelly, 2017), its waste management, and particularly its recyclability (Heidbreder et al., 2019). For many of these issues, knowledge about specific flows of plastics in society is necessary to start a discussion and decide on measures to combat pollution and increase the circularity of plastics. Depending on the life-cycles of the various polymers and the products made out of those plastics, the sizes of material flows and their release pathways can differ significantly (Kawecki and Nowack, 2019; Kawecki et al., 2018). Single-use products, for example, have very different life-cycles, release potentials, and recyclabilities compared to materials incorporated into building structures.

Material Flow Analysis (MFA) is a scientific modeling approach for quantifying the flows of elements, compounds, and materials through the anthroposphere (Brunner and Rechberger, 2004). MFA has been

used for many years to analyze the flows of plastics through society; however, this has usually been done without distinguishing between individual polymers (Bogucka et al., 2008; Joosten et al., 2000; Mutha et al., 2006; Patel et al., 1998; Van Eygen et al., 2017). Geyer et al. (2017) used a dynamic approach to quantify the “production, use and fate of all plastics ever made”, but due to their global scope and the associated uncertainty, these results are only of limited value for smaller geographical areas and polymer-specific evaluations.

MFAs of a specific polymer have often been performed for PVC because of its potential health impacts (Ciacci et al., 2017; Nakamura et al., 2009; Tukker et al., 1997). Dynamic MFAs, with a defined system boundary, have been performed for PVC in China (Liu et al., 2020; Zhou et al., 2013), Japan (Nakamura et al., 2009), and Europe (Ciacci et al., 2017). Considering product lifetimes, combined with time-dependent trade- and transfer-flows, dynamic MFA enables the identification and quantification of the polymer's accumulation in different societal stocks and sinks. Jiang et al. (2020) used a dynamic model to assess the stocks and flows of five different polymers in China from 1978 to 2017. However, their model considered just six different product sectors and

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could thus only model a generic life-cycle. A dynamic MFA model for PE, PP, and PET in Europe was recently published (Eriksen et al., 2020), but it only considered eight generic product sectors. A static MFA was made for 15 different polymers in the USA, but it also used just eight product sectors (Heller et al., 2020). Other polymer-specific MFAs have been performed for PET because of its high potential for recycling (Kuczenski and Geyer, 2010). Wang et al. (2020), for example, have developed a dynamic model for PET bottles in China, focusing exclusively on its specific use in bottles and excluding all other uses, such as in other packing applications or textiles.

A separate analysis of the flows of seven important polymers within the EU (Kawecki et al., 2018) looked at low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP), polystyrene (PS), expanded polystyrene (EPS), polyvinylchloride (PVC), and polyethylene terephthalate (PET). This work formed the basis of a quantification of these polymers' flows to the environment (Kawecki and Nowack, 2019). These authors categorized each polymer's flows through nine product sectors and 35 product categories and then on into ten waste collection systems. This model provided a very detailed description of the life-cycle of these seven polymers. However, the model used a static MFA approach and, thus, no stocks or sinks could be quantified.

The textile sector constitutes a major end-user for several polymers, e.g., PET, PP, and polyamide. However, in many reports about plastic production and use, e.g., from PlasticsEurope (PlasticsEurope, 2018), fibers are excluded because the fiber sector is organized across different trade organizations. Therefore, many MFA studies did not assess fibers, e.g. Jiang et al. (2020). By combining data from the plastics and fiber industries, Kawecki et al. (2018) provided polymer-specific mass-flow data that also included fiber production and usage. For some polymers, such as PET and PP, fibers represent a sizeable fraction of their use, e.g., textile manufacturing in Europe uses 1800 kT of PET compared to 3900 kT used in non-textile manufacturing.

Although plastic is omnipresent in our society and concerns about its impacts on human health and the environment are rising, not much is known about polymer-specific mass flows through our society, as mentioned above. Polymer-specific modeling is important as distinctions between different polymers is needed for exposure assessments (Kawecki and Nowack, 2019). Environmental or human risk assessment should ideally distinguish individual materials since toxicities may differ depending on the material itself and the additives included. Individual polymers' release-pathways to the environment are very different and depend on those polymers' life-cycles (Kawecki and Nowack, 2019). Furthermore, material and life-cycle stages must be distinguished in order to incorporate released plastic materials into Life Cycle Inventories (LCIs). Also, to better understand the recycling systems that normally target specific polymers, we need to know more about individual polymer flows.

Whereas all MFA studies separate the use phase into different product sectors, the number of specific product categories is usually very limited. Detailed knowledge about product categories is indispensable when aiming to use that data to improve recycling schemes or predict the environmental release of plastics. For example, in the packaging sector, large differences in disposal, recycling, and environmental release exist between different types of packaging, e.g., consumer bags and non-consumer packaging. Whereas consumer bags are prone to littering due to their potential use outdoors, the films used to package durable goods are discarded in a much more controlled way. There are also very large differences in the recycling potential between different packaging types, e.g., PET bottles and the PET used in other packaging applications. In light of this, there is a clear need for MFA studies that split polymer flows into product sectors and more specific product categories.

Therefore, this work's goal was to provide a basis for future analyses of impacts linked to specific polymers. Thus, based on the Dynamic Probabilistic Material Flow Analysis (DPMFA) method (Bornhöft et al.,

2016), we built a DPMFA model including all the life-cycle phases for seven commodity thermoplastics. The model was created for Europe, with a temporal scale for production and trade since 1950, and it considered the in-use stocks resulting from varying product lifetimes. Each polymer's flows were split into nine product sectors and 35 product categories, based on Kawecki et al. (2018), and they included fiber uses. This article is an extension of the previously published article by Kawecki et al. (2018), in which the polymer flows through society were modelled for a single year, omitting stocks and other time-dependent factors.

2. Methods

Material Flow Analysis (MFA) is the common approach to modelling flows and stocks of materials through a specified portion of the economy (Brunner and Rechberger, 2004), and different software products exist for performing it. The present work was carried out using the DPMFA Python package (Bornhöft et al., 2016), a dynamic and probabilistic version of MFA that allows uncertainties in all the model's parameters to be incorporated as probability distributions. This work also used an update of the dynamic MFA developed by Rajkovic et al. (2020), and the DPMFA package was developed further by migrating from Python 2.7 to Python 3 and implementing an automatic model setup from SQL databases.

2.1. System boundary

This study considered seven different thermoplastics, chosen based on their frequency of use (PlasticsEurope, 2018) and presence in the environment (Gasperi et al., 2014; Sadri and Thompson, 2014): LDPE, HDPE, PP, PS, EPS, PVC, and PET. Fibre and textile applications fell within the material definitions used and are modelled distinctly. Additives were removed from the total plastic material for a coherent overview of every life-cycle stage (Kawecki et al., 2018).

The system includes the complete life-cycle, with a strong focus on a detailed description of the consumption stage (Fig. 1). The model's structure is essentially identical to the structure used previously by Kawecki et al. (2018), except for the inclusion of stocks of the various product categories. The geographical boundary was set to Europe, defined as the EU-28 plus Norway and Switzerland, and modeled as a single entity without national variations. Flows were modeled for the period from 1950 to 2016. The model's structure can be split into five stages: production, manufacturing, consumption, waste collection, and waste treatment. A distinction was made between primary and secondary (recycled) production. A total of 35 product categories were considered, of which 10 were textile applications. Recycling was split into two steps: the collection of various waste streams and the recycling itself. A certain fraction of each collection and recycling process may flow to landfill and incineration, either lost to collection or as waste from the recycling process.

2.2. The DPMFA package

A simple MFA can be described mathematically as depending on two mathematical objects: the transfer coefficient matrix, including all the transfers from one compartment to the next, and an inflow or input vector, which describes the mass introduced into each compartment across the system boundary. The DPMFA package was thoroughly described in the study that introduced it (Bornhöft et al., 2016), and this section only gives a short overview of it. One critical development, contributed by a later publication (Rajkovic et al., 2020), was also used here. A new class of model compartments was created, for which outflows are not based on delays (as in the original DPMFA model) but vary from one period to another. The package's two essential characteristics of dynamism and Bayesian aspects distinguish it from simple MFA methods.

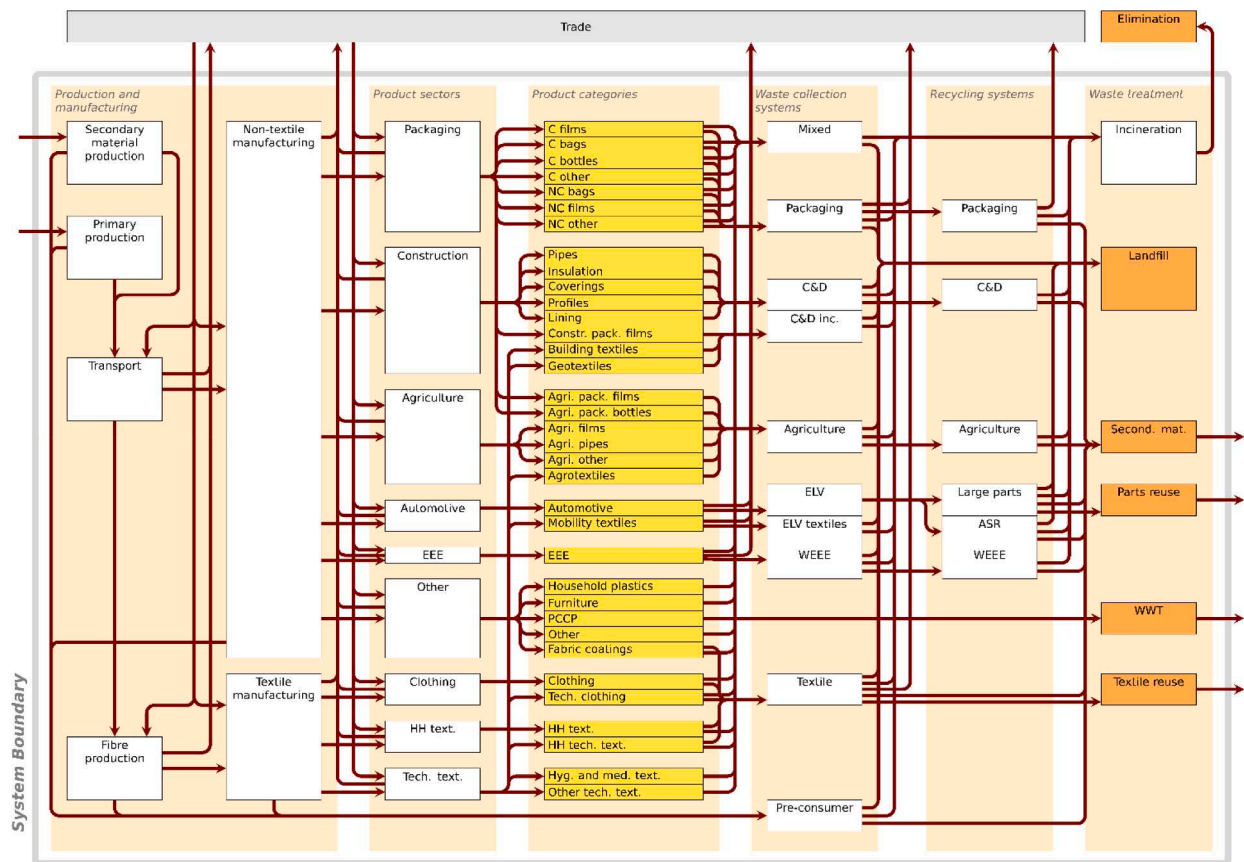


Fig. 1. System boundary for the material flow analysis. The diagram was adapted from (Kawecki et al., 2018). Stocks are shown in yellow and sinks in orange. Trade compartment data determines import flows into the system and export flows as a sink. EEE: electrical and electronic equipment; PCCP: Personal care and cosmetics products; HH: household; C&D: construction and demolition; ELV: end-of-life vehicles; WEEE: waste electrical and electronic equipment; ASR: auto-shredder residues; WWT: wastewater treatment.

The dynamic aspect introduces a temporal dimension:

- including dynamic inflows into the system,
- using time-dependent transfer coefficients (TCs), which describe the mass allocation from one process to another, and
- calculating the accumulation of product stocks due to different product lifetimes and delayed product waste generation.

The temporal dimension is described as a succession of time increments—in our particular case, years.

The Bayesian aspect introduces a probability distribution describing the uncertainty in every parameter in the model. Two mathematical objects are formulated: an input vector describing the starting mass in every compartment and a TC matrix. These objects are then used to solve a matrix Eq. (10)⁶ times in a Monte Carlo setting, yielding Bayesian distributions of the masses contained in each compartment. The shape of the Bayesian distributions used in this study depended on data availability (Gottschalk and Nowack, 2013) and quality (Laner et al., 2016), as described by Kawecki et al. (2018). These distributions can take a triangular or trapezoidal form, depending on whether one or two data points are available in the literature. In addition, a coefficient of variation is associated with each data point obtained from the literature (Kawecki et al., 2018; Laner et al., 2016) based on a pedigree matrix (see Table S1 in the Supporting Information) with five different data quality indicators. All the probability distributions were truncated below 0. The TC probability distributions were truncated above 1 to ensure mass conservation.

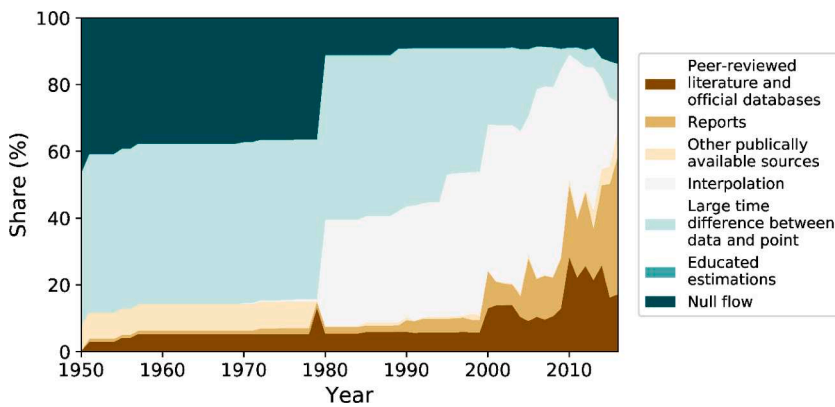
2.3. Improvements to the DPMFA package

The DPMFA Python package described above was updated throughout this work to improve its reproducibility and usability. First, a few previously identified bugs were fixed and regression tests (Rothermel and Harrold, 1994) were added. Bugs included failing to include time-dependent transfers in one compartment and other corner cases. Second, early validity checks and debug methods were added, such as variable type-checking helpers that return human-readable warning and error messages. Finally, the codebase was updated to be compatible with the newer Python 3.8 version while maintaining backwards compatibility.

A further improvement to the DPMFA package was automating the package and the simulation set up process, i.e., implementing the conceptual model in Python code and separating it from the instantiation of simulation data. This automation was done by providing the data in Excel tables and letting the new software discover, based on their names, which unique compartments and flows were defined in that data. Based on this, an SQL database was created using SQLite in Python. The Python scripts and SQL database used to perform the calculations are available at <https://github.com/empa-tsl/plastic-dpmfa>.

2.4. Data collection

Data were obtained from (in order of priority) official databases, peer-reviewed publications, expert opinions, gray literature, and websites (Fig. 2). The data collection approach closely resembled that used by Kawecki et al. (2018), with the addition of a temporal dimension. Complete time-series of inflows and transfers were sought, but little data



was found for the years before 2000. Most of the high-quality data came from recent years, with quality steadily decreasing from the mid-2010s to 2000 (Fig. 2). The small increase in the share for literature in 1979 is based on the assessment that before 1980, recycling and incineration were negligible (Geyer et al., 2017). Where time-series data points were missing from the literature, data were either interpolated linearly between existing estimates or set equal to the nearest data point if interpolation was impossible. For a complete description of the data included in the model, see the SQL database and additional descriptions provided in the Supporting Information.

The time series for the primary production of the seven polymers were constructed using data from industrial reports (AMI, 2015; PlasticsEurope, 2004, 2005, 2013, 2015a, b, 2016, 2017, 2018) and Eurostat (Eurostat). Estimates for secondary production assumed that no recycling took place before 1972 (American Chemistry Council), used data available in the literature (PlasticsEurope, 2008; Simon and Hupfer, 2015), and interpolated missing data. See the Supporting Information for complete descriptions of both.

All trade data were obtained using the method described by Kawecki et al. (2018), based on data from the Eurostat database (<https://ec.europa.eu/eurostat/data/database>). All the goods in the database suspected of containing plastic were examined, and the shares of individual polymers within those goods were estimated using additional information from the literature. The shares of the polymers for the categories of goods considered and the literature used can be found as Supporting Information in Kawecki et al. (2018). The least well-known trade flow in their assessment was the packaging around traded goods. A separate calculation had to be performed for this flow, as validated in Kawecki et al. (2018) and explained in detail in their Supporting Information. Eurostat trade data was available from 2000 onwards, except for rare instances where data only became available later. We assumed that traded amounts were negligible in 1950, and missing data between 1950 and 2000 were interpolated. Trade was only considered for the following processes in the system: transport, fiber production, non-textile manufacturing, textile manufacturing, packaging, automotive, electrical and electronic equipment (EEE), clothing, household textiles, and technical textiles. Positive net imports (imports minus exports) were treated as an external inflow, and negative net imports (i.e., net exports) were instead used to calculate TCs from their respective processes.

The TCs for the distribution of plastic from non-textile manufacturing to the product sectors were kept constant throughout the modeling because of a lack of data (Simon et al., 2016). The probability distributions are nevertheless not identical because of the uncertainty associated with the time difference. For the TCs from textile manufacturing, data were available for 1990 (Hatch, 1993) and 2012 (Wazir Management Consultants, 2014), and the remaining TCs were interpolated. For the distributions from product sectors to product categories, enough data were found to create time series for technical

Fig. 2. Origin of the data for all years, combining all transfer coefficients and inputs for all seven materials. Data in the “Large time difference between data and point” group are for data where there is ten years or more time difference between the data obtained from the literature and the data modeled. “Interpolation” data also includes cases using the nearest existing data point. “Null flow” data were based on literature stating that no such flows existed either before a certain date or for certain materials. “Educated estimations” data for non-zero flows only includes cases where no data could be found; representing less than 0.2% of the total, they are invisible on the chart. Flows defined as the remaining fraction of a mass exiting a compartment are excluded from the figure.

textiles, PCCP, and some PVC products. The remaining TCs were constant throughout the period, although uncertainty varied.

Lifetime distributions were used to model a specific product category’s waste generation rate. A detailed description of all the assumptions required to create lifetime distributions for each of the 35 product categories is given in Table S6 of the Supporting Information.

We assumed that no recycling took place until 1980 (Geyer et al., 2017). Data on waste collection and recycling were available for more recent years in the literature (Delgado et al., 2007; Huisman et al., 2015) and databases (Eurostat), and the missing data were interpolated. Full citations are available in the Supporting Information. Release into wastewater was included for down-the-drain products such as shampoos that contain primary microplastics and where complete release into wastewater is part of the normal and intended life-cycle.

2.5. Uncertainty analysis

After the simulation, 10,000 values are obtained for each result (flows and masses in sinks and stocks), which form the Bayesian probability distributions in the DPMFA. For each distribution, a relative uncertainty can be calculated to compare flows with different magnitudes. The relative uncertainty was calculated as follows:

$$\Delta_{Rel} = \frac{SD}{Mean} \cdot 100$$

where SD stands for the standard deviation, and Mean for the mean of the distribution. The result is expressed in percent.

3. Results

3.1. Probability distributions

Due to the MFA model’s fully probabilistic nature, all the results obtained were in the form of probability distributions that could then be analyzed statistically. The link to download the complete results is given at the end of the manuscript. Four examples of these distributions are presented in Fig. 3. Fig. 3a shows the probability distribution of European production of the seven polymers. The triangular distribution that was used to consider production uncertainty is visible. The next two panels present two intermediate results: Fig. 3b shows LDPE flows into the product category of “Consumer films” for 1970, 1990 and 2010. Fig. 3c presents PET flows from clothing to mixed waste for the same years. Compared to Fig. 3a, the distributions are broader and smoother because their results were influenced by many different probability distributions. Fig. 3d shows the stock of polymer mass of our seven different polymers stored as electrical and electronic equipment in 2016. In the following sections, most of the figures only show mean values in order to simplify them, but full probability curves can be extracted from all the model’s results if needed.

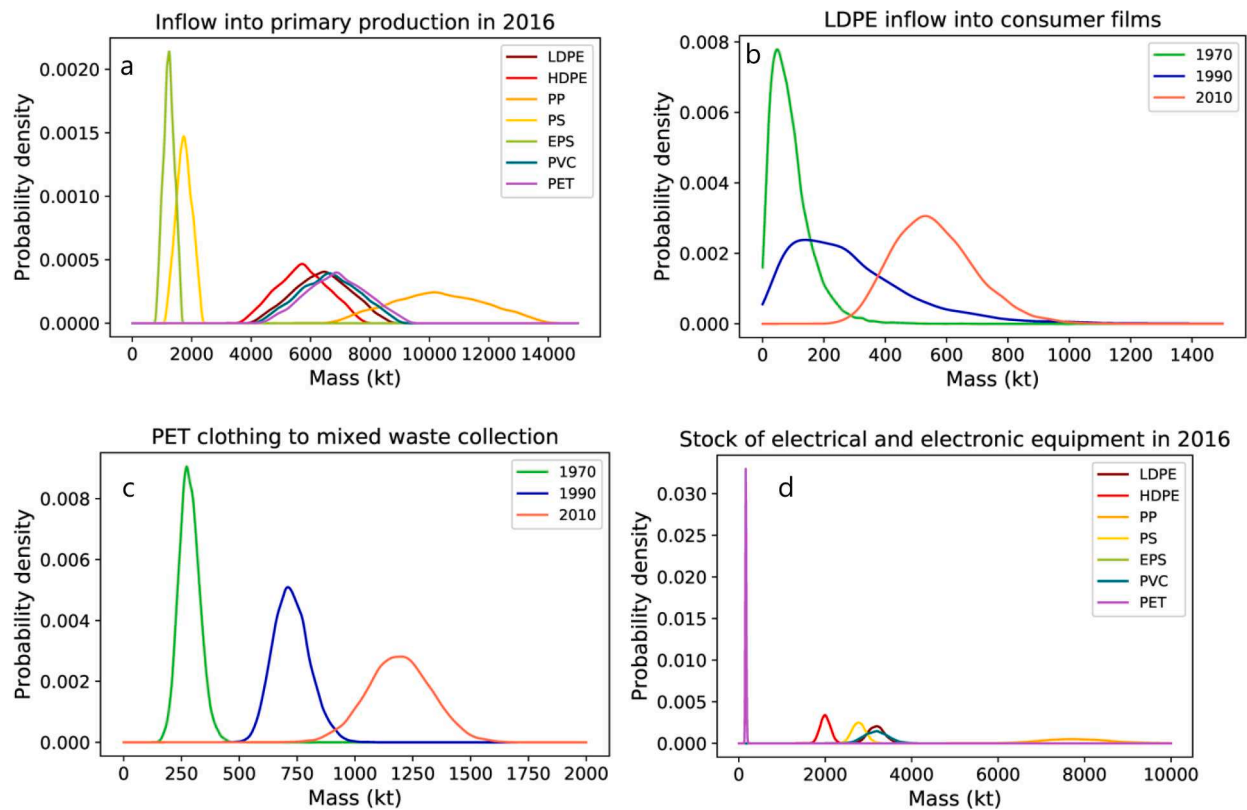


Fig. 3. Selected probability distributions from the results of the Monte Carlo simulation. Examples are shown for (a) primary production inputs for the seven polymers, (b) LDPE flows into consumer films, (c) PET flows from clothing to mixed waste collection, and (d) the mass of the seven polymers accumulated in stocks of electrical and electronic equipment.

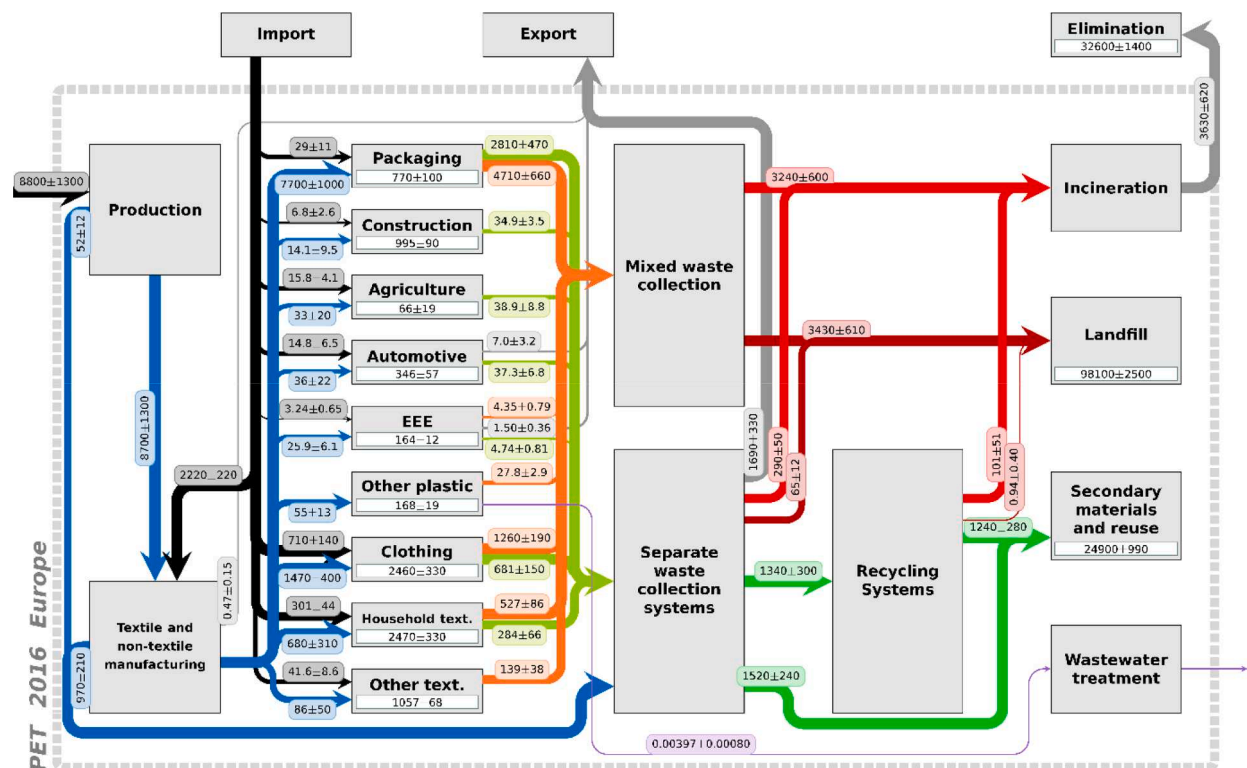


Fig. 4. Simplified flowchart showing PET flows, stocks, and sinks in 2016. All units are in thousand metric tonnes (kt). The means and standard deviations of the probability distributions are shown, both rounded to two significant figures of the standard deviation. Flow widths are larger for larger flows, and colors are for visualization only. The products displayed are combinations of the product category groups from Fig. 1, i.e., "Packaging" is the sum of seven product categories and "Agriculture" is the sum of six.

3.2. MFA diagrams

Fig. 4 shows the flows, stocks, and sinks for PET in 2016. This figure aggregates flows and compartments to help visualization. European production made up 90% of the PET used in Europe in 2016, with only 10% imported from abroad as primary forms or products. Most of the inflow in consumption flowed to packaging, at 7700 ± 1000 kt, and to clothing, at 1470 ± 400 kt. The largest stocks were found for textiles: 2460 ± 330 kt for clothing and 2470 ± 330 kt for household textiles. Ten percent of packaging applications remained in stock for one year because of the chosen lifetime distributions (Delgado et al., 2007; Schelker and Geisselhardt, 2010). The stocks of PET in construction, agriculture, and the automotive sector may be larger than first expected because of the inclusion of technical textiles in the definition of the aggregated products. Forty-two percent of PET waste was collected separately; the rest was collected as mixed waste and incinerated or landfilled. From 1950 to 2016, totals of $32,600 \pm 600$ kt of PET were incinerated, $98,100 \pm 2500$ kt were landfilled, and $24,900 \pm 990$ kt were reused or recycled.

3.3. Time series

The diagrams in Fig. 5 show the development of the production, consumption, and consumption in-use stocks of our seven polymers over time, plus their combined total final sinks. Being a fully dynamic model, the flows of the seven polymers were tracked individually from 1950. Primary production of the seven polymers in Europe (Fig. 5a), plus imports and reused materials, constitute the only inputs of polymers into the system. Consumption (Fig. 5b) is calculated from all the flows that lead into the 35 product categories considered. These flows also include material from recycled and reused materials. Polymer quantities that are exported directly after production and manufacturing, as well as pre-consumer waste, are not included in consumption. Over the entire period, PP has remained the polymer with the highest production and

use. Recently, PET has surpassed HDPE and LDPE due to very rapid production growth in the last five years. Other polymers, such as LDPE, PVC, and PS, have exhibited stable consumption since 2005, whereas EPS has shown a steady decline in the last years. The accumulation of in-use stock—i.e., stored in polymer-containing applications during the use-phase—shows very different behavior to the development of consumption (Fig. 5c). Behavior is now determined by product lifetimes during use, and the details are presented below when analyzing the in-use stock for 2016 in more detail. The evolution of the model's sinks is presented in Fig. 5d. The amounts stored in landfills are shown together with the masses eliminated, reused, and exported. The model considers flows into wastewater during the use-phase, but these are not visible in the figure. A detailed analysis of the polymers contained in the in-use stock is made below.

Fig. 6 shows the consumption for each of the seven polymers by product sector. All polymers display an increase in consumption since 1950, also visible in the total consumption shown in Fig. 4b. The seven polymers' product sector profiles look very different depending on their typical uses. Packaging was a major use for LDPE and HDPE over the whole period, and in recent years packaging has been the driving force for the vast increase in PET consumption. PP packaging has also increased in use over the last 15 years. LDPE and HDPE packaging have not expanded since around 2005. Construction is the most important sector for EPS and PVC use. The decrease in EPS consumption in recent years can mainly be explained by the decrease in its use in construction EPS. Clothing and household textiles are also relevant to PET use and, to a lesser extent, PP use. Given the very limited availability of historical data on product sector distributions, the model could only consider limited changes in polymer product distributions over time.

3.4. Consumption and stock magnitudes

Each product sector was split into the various product categories that form the core of the DPMFA model. Product categories have diverse life-

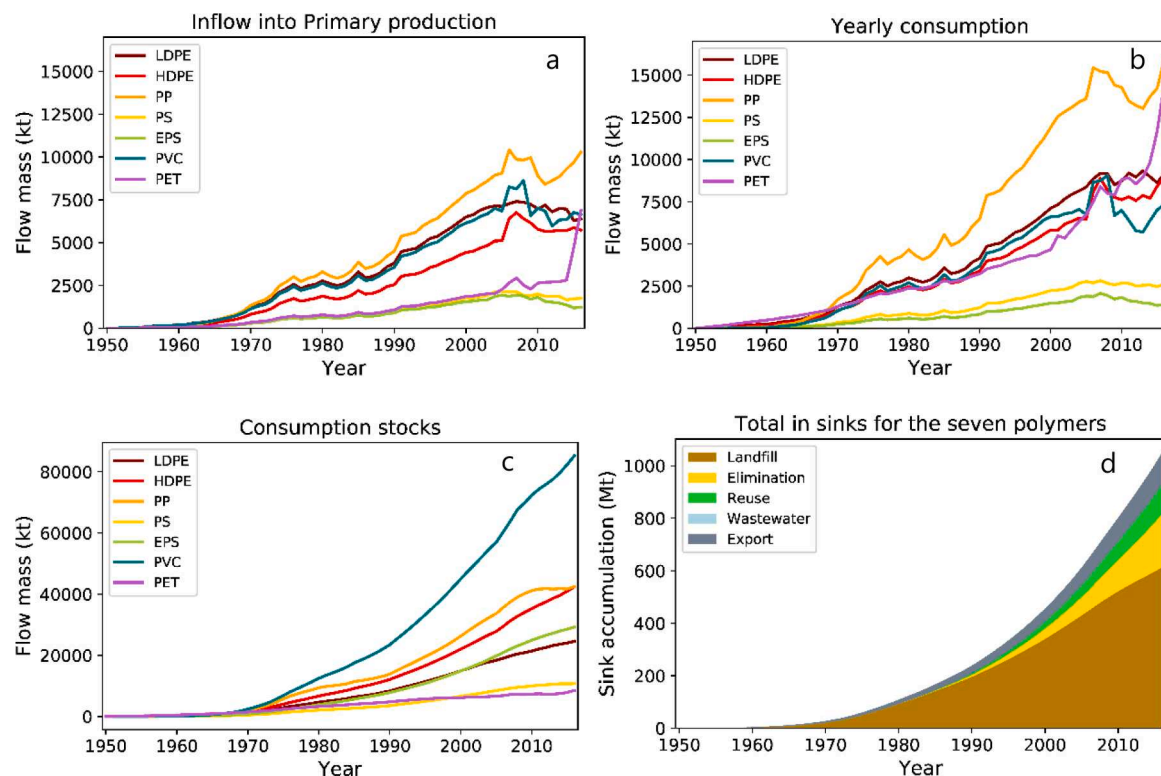


Fig. 5. Time series from 1950 to 2016 for (a) primary production, (b) consumption, (c) amounts contained in the in-use stock, and (d) combined amounts of the seven polymers analyzed polymers in final sinks for LDPE, HDPE, PP, PS, EPS, PVC and PET. These figures show the means of the probability distributions.

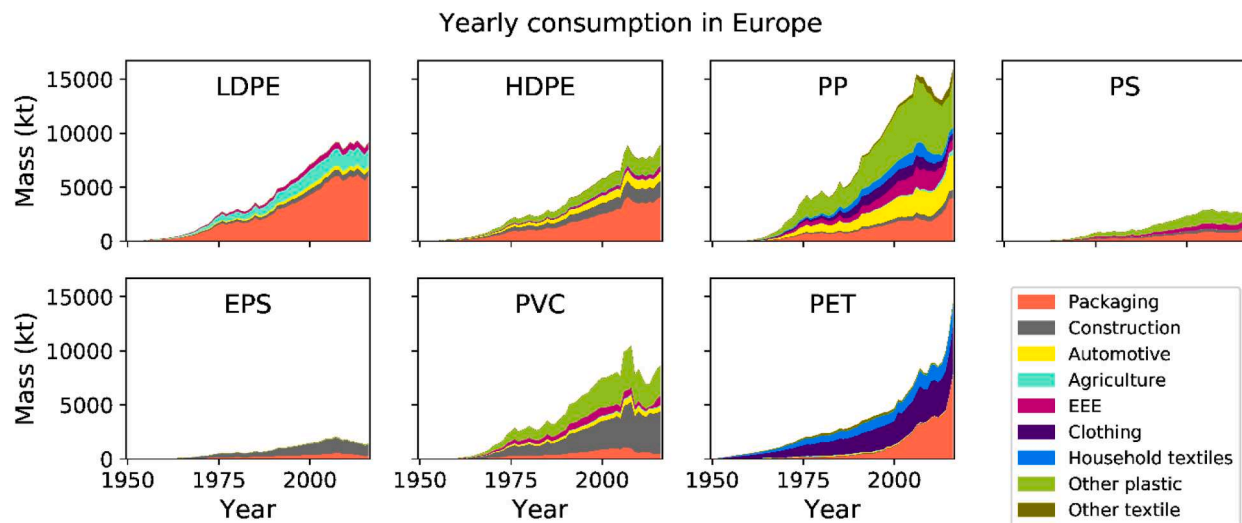


Fig. 6. Changes in LDPE, HDPE, PP, PS, EPS, PVC, and PET consumption in Europe from 1950 to 2016, by material and sector. Graphs show the means of the probability distributions. EEE: electrical and electronic equipment.

cycles and different fates within the system. On its left side, Fig. 7 shows the various polymers' contributions to the product categories in 2016. The seven polymers' major applications were in various packaging and construction applications, the most important being "Other consumer packaging", "Consumer bottles", "Other non-consumer films", "Pipes and ducts", "Other non-consumer packaging", and "Consumer bags".

LDPE and HDPE dominated most of the film and packaging applications, including "Other non-consumer films" like "Agricultural packaging films" and "Building packaging films". PET and PP shared most of the textile applications, with PP dominating most of the technical applications and PET dominating the remainder.

The right half of Fig. 7 shows the magnitudes of 2016's in-use stocks

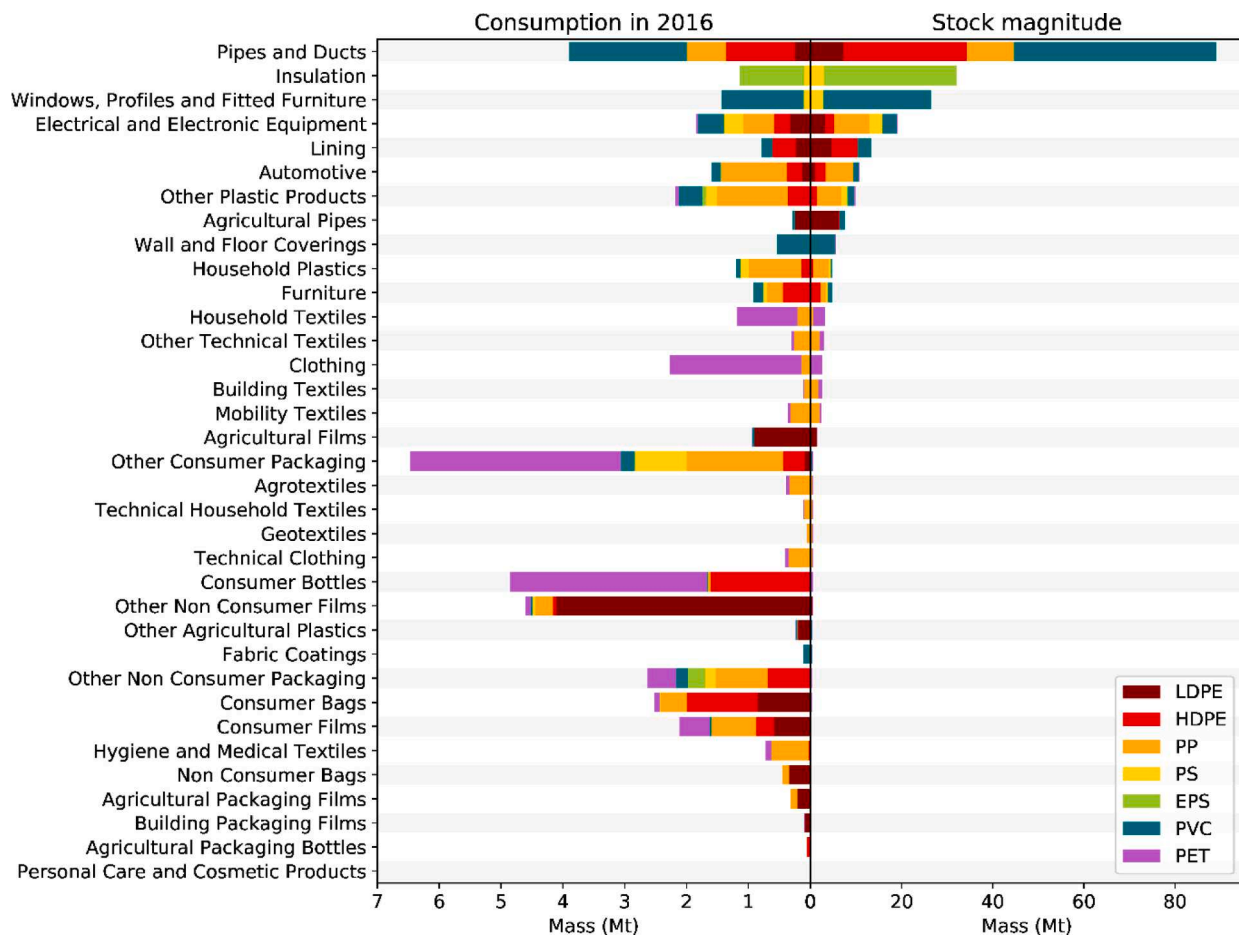


Fig. 7. Overview of consumption in 2016 in Mt/year (left side), and the magnitude of stocks (right side) by product category and material in Mt accumulated from 1950 to 2016. The chart shows the means of the probability distributions.

for the different product categories, separated into the various polymers. Although packaging dominated consumption, the construction sector dominated the in-use stocks of plastic polymers. Packaging applications were held in relatively small stocks of the polymers analyzed polymers, and they are barely visible on the chart at this scale. This is due to packaging's very fast turnover compared to that of the plastics used in construction and locked into buildings and other infrastructure for decades. The biggest stocks were found for pipes and ducts, insulation, and windows. Other significant product categories were electrical and electronic equipment and automotive applications, which also have long lifetimes compared to packaging applications. The main polymers in the in-use stock were PVC, EPS, and HDPE.

3.5. Polymers' final whereabouts

The final compartments considered by the model were elimination, landfill, recycling and reuse, export, and direct release into wastewater during use. Fig. 8 shows the cumulative amount of the seven polymers in these stocks between 1950 and 2016, in addition to the amount stored in the in-use stock (light blue). The wastewater compartment is not shown here because it is too small to be visible at this scale. It is important to understand that this release to wastewater only includes intended release during the use of down-the-drain personal care products. The unintended release of polymer fibers during the washing of textiles was not included in this MFA study, but it was considered in the second step of modeling when all flows to the environment were quantified (Kawecki and Nowack, 2019). There were very large differences between the polymers. Thus, the major final compartment for LDPE, HDPE, PP, PS, and PET was landfill (48%–60%). The largest fraction for EPS and PVC was in the in-use stock (51% and 39%, respectively). Elimination—the incineration and complete destruction of the polymers in waste incineration plants—had shares of 15%–19% for LDPE, HDPE, PP, PS, and PET, and 10% for EPS and PVC. The recycled fraction was between 5% for PS and 14% for PET. Exports of LDPE and HDPE were very small (3%–5%) and reached up to 16% for PVC. Between 1950 and 2016, 69% of PVC exports originated from production. For 2016, this fraction went up to 88%. In comparison, almost no net exports of LDPE and HDPE in their primary forms took place between 1950 and 2016. The present model only considered net trade (import–export) from trade databases, which means that there may only be one import or export flow from a given compartment.

Table 1 presents the absolute values for the data shown in Fig. 8 in relative units. Additionally, the per-capita amounts for the different polymers in the various stocks and sinks are shown. The consumption of the seven polymers considered was 90 ± 5 kg/cap in 2016. The total weight of all the polymers in the in-use stock in 2016 was 465 ± 11 kg/cap. The overall mass of the seven polymers in all stocks and sinks was 1.3 billion tons, yielding 2469 ± 39 kg per person living in Europe. Overall consumption of the different polymers was PP (591 kg/cap), LDPE (510 kg/cap), PVC (422 kg/cap), HDPE (372 kg/cap), PET (339 kg/cap), PS (120 kg/cap), and EPS (102 kg/cap). The total in-use stock of all seven polymers was 856 million tons. The total weight in landfills was 1260 kg/cap, in order: LDPE (308 kg/cap), PP (295 kg/cap), PVC (203 kg/cap), HDPE 8199 kg/cap), PET (188 kg/cap), PS (65 kg/cap), and EPS (63 kg/cap).

3.6. Recycling

Fig. 9 shows the recycling rates for the seven polymers between 1980 and 2016. All the rates have grown continuously, reaching values between 33% for PET, 20%–24% for HDPE, LDPE, EPS, and PVC, and 11%–14% for PS and PP. The most notable increase was observed for PET, which has surpassed all the other polymer types in the past few years. The right side of Fig. 9 shows a comparison of the calculated recycling inflows and outflows for each polymer. Inflows were extracted from available data on the weights of polymers reused and re-injected into the market each year. Outflows were determined by modeling the consumption, lifetimes, and recycling rates per waste stream. For most polymers, the agreement between the two approaches was very good; only PVC had a large disagreement. PVC inflow was determined using data from the VinylPlus program alone (VinylPlus, 2020), and these were only available as of 2001. PVC outflow was calculated using the same method as for the other polymers. The good agreement between these two independent assessments for the various polymers added confidence about the magnitudes of the recycling rates—values that are of great interest in the context of a circular economy.

3.7. Uncertainty assessment

Since this study relied on Bayesian modeling, a probability distribution was associated with each parameter in the system. Each distribution was defined using data from the literature as central values and

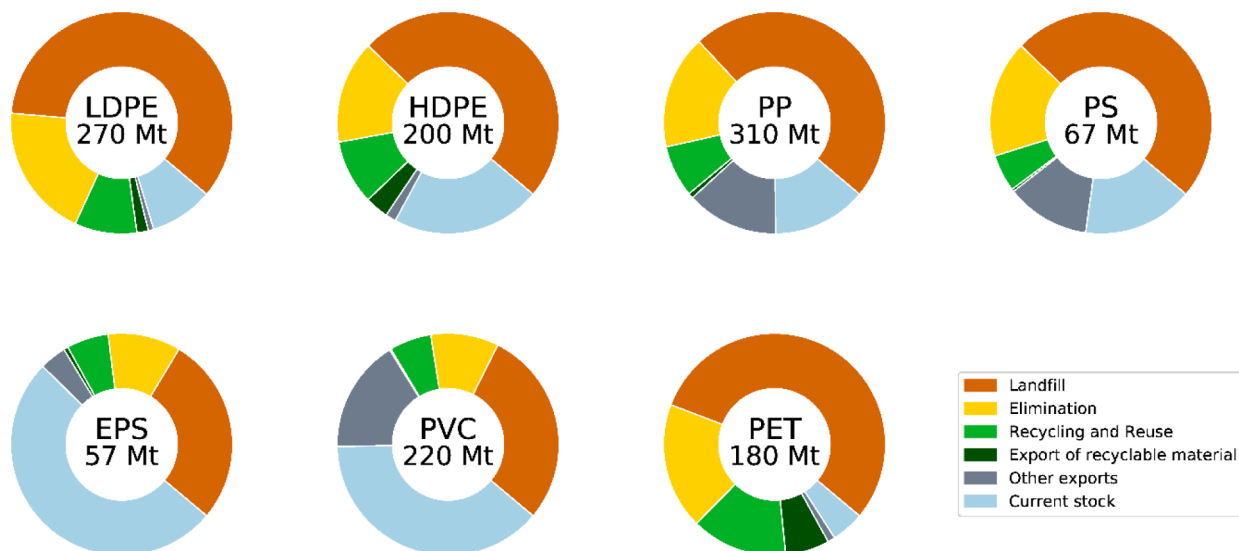


Fig. 8. Overview of the whereabouts of all the plastic consumed in Europe since 1950 in 2016. The charts show the means of the probability distributions. The pie chart centers show the total amount of each material put on the market since 1950. The data corresponding to this figure can be found in Table 1. Data corresponding to the “Wastewater” sink were omitted since they are too small to be visible at this scale—they can be found in Table S7.

Table 1

Consumption in 2016 and quantities in stocks and sinks in 2016 for the seven different polymers. Numbers in black are in thousands of tonnes, and numbers in gray are in kilograms per capita based on Eurostat's population data for 2016 (Eurostat, 2020). The table shows the means and standard deviations of the probability distributions. Standard deviations were rounded to two significant figures, and the mean's precision was adapted to the standard deviation. Because of this rounding, sums may not correspond exactly to the totals displayed.

	LDPE	HDPE	PP	PS	EPS	PVC	PET	Total
Consumption in 2016	8600 ± 1000 kt 16.4 ± 2.0 kg/cap	7200 ± 890 kt 13.7 ± 1.7 kg/cap	11,000 ± 1500 kt 20.9 ± 2.8 kg/cap	1940 ± 260 kt 3.71 ± 0.50 kg/cap	1370 ± 190 kt 2.62 ± 0.36 kg/cap	5870 ± 810 kt 11.21 ± 1.55 kg/cap	11,200 ± 1200 kt 21.4 ± 2.3 kg/cap	47,100 ± 2500 kt 90.0 ± 4.8 kg/cap
In-use stock	24,600 ± 1500 kt 47.0 ± 2.8 kg/cap	42,400 ± 2300 kt 81.0 ± 4.4 kg/cap	42,500 ± 2200 kt 81.1 ± 4.2 kg/cap	10,740 ± 580 kt 20.5 ± 1.1 kg/cap	29,200 ± 1800 kt 55.8 ± 3.3 kg/cap	85,300 ± 4200 kt 162.9 ± 8.0 kg/cap	8500 ± 390 kt 16.23 ± 0.74 kg/cap	243,300 ± 5700 kt 465 ± 11 kg/cap
Elimination	51,900 ± 2300 kt 99.1 ± 4.5 kg/cap	29,200 ± 1400 kt 55.7 ± 2.7 kg/cap	51,200 ± 2600 kt 97.7 ± 5.0 kg/cap	11,320 ± 650 kt 21.6 ± 1.2 kg/cap	6030 ± 390 kt 11.5 ± 0.7 kg/cap	21,900 ± 1300 kt 41.8 ± 2.6 kg/cap	32,600 ± 1400 kt 62.2 ± 2.6 kg/cap	204,100 ± 4300 kt 389.7 ± 8.1 kg/cap
Landfill	159,100 ± 7000 kt 304 ± 13 kg/cap	95,300 ± 4400 kt 182.1 ± 8.3 kg/cap	148,800 ± 7400 kt 284 ± 14 kg/cap	32,600 ± 1800 kt 62.2 ± 3.4 kg/cap	15,700 ± 1000 kt 29.9 ± 2.0 kg/cap	63,600 ± 3800 kt 121.4 ± 7.3 kg/cap	98,100 ± 2500 kt 187.4 ± 4.8 kg/cap	613,000 ± 12,000 kt 1171 ± 23 kg/cap
WWTP	0.0 ± 0.0 kt 0.0 ± 0.0 kg/cap	86.7 ± 7.1 kt 0.166 ± 0.014 kg/cap	4.97 ± 0.44 kt 0.00950 ± 0.00084 kg/cap	0.0 ± 0.0 kt 0.0 ± 0.0 kg/cap	0.0 ± 0.0 kt 0.0 ± 0.0 kg/cap	0.0 ± 0.0 kt 0.0 ± 0.0 kg/cap	0.266 ± 0.032 kt 0.000508 ± 0.000061 kg/cap	91.94 ± 7.1 kt 0.176 ± 0.014 kg/cap
Recycling and reuse	24,100 ± 1500 kt 46.1 ± 2.8 kg/cap	18,300 ± 1000 kt 35.0 ± 1.9 kg/cap	23,300 ± 1300 kt 44.5 ± 2.6 kg/cap	3600 ± 280 kt 6.88 ± 0.53 kg/cap	3480 ± 310 kt 6.65 ± 0.59 kg/cap	13,490 ± 950 kt 25.8 ± 1.8 kg/cap	24,900 ± 990 kt 47.5 ± 1.9 kg/cap	111,300 ± 2600 kt 212.5 ± 5.0 kg/cap
Export of recycled material	4430 ± 430 kt 8.46 ± 0.83 kg/cap	6640 ± 570 kt 12.7 ± 1.1 kg/cap	2330 ± 160 kt 4.45 ± 0.30 kg/cap	246 ± 25 kt 0.470 ± 0.048 kg/cap	374 ± 57 kt 0.72 ± 0.11 kg/cap	470 ± 46 kt 0.898 ± 0.087 kg/cap	11,360 ± 650 kt 21.7 ± 1.2 kg/cap	25,850 ± 980 kt 49.4 ± 1.9 kg/cap
Other exports	2060 ± 160 kt 3.93 ± 0.31 kg/cap	2990 ± 190 kt 5.71 ± 0.37 kg/cap	41,500 ± 2000 kt 79.2 ± 3.9 kg/cap	8120 ± 410 kt 15.51 ± 0.79 kg/cap	2210 ± 130 kt 4.22 ± 0.25 kg/cap	36,500 ± 1600 kt 69.68 ± 3.10 kg/cap	1910 ± 130 kt 3.65 ± 0.24 kg/cap	95,300 ± 2600 kt 181.9 ± 5.0 kg/cap
Sum of all stocks and sinks (excl. consumption in 2016)	266,000 ± 10,000 kt 510 ± 20 kg/cap	195,000 ± 7200 kt 372 ± 14 kg/cap	310,000 ± 13,000 kt 591 ± 24 kg/cap	66,600 ± 3200 kt 127.2 ± 6.0 kg/cap	57,000 ± 2800 kt 108.9 ± 5.3 kg/cap	221,200 ± 9700 kt 422 ± 18 kg/cap	177,400 ± 4100 kt 338.7 ± 7.8 kg/cap	1,293,000 ± 21,000 kt 2469 ± 39 kg/cap

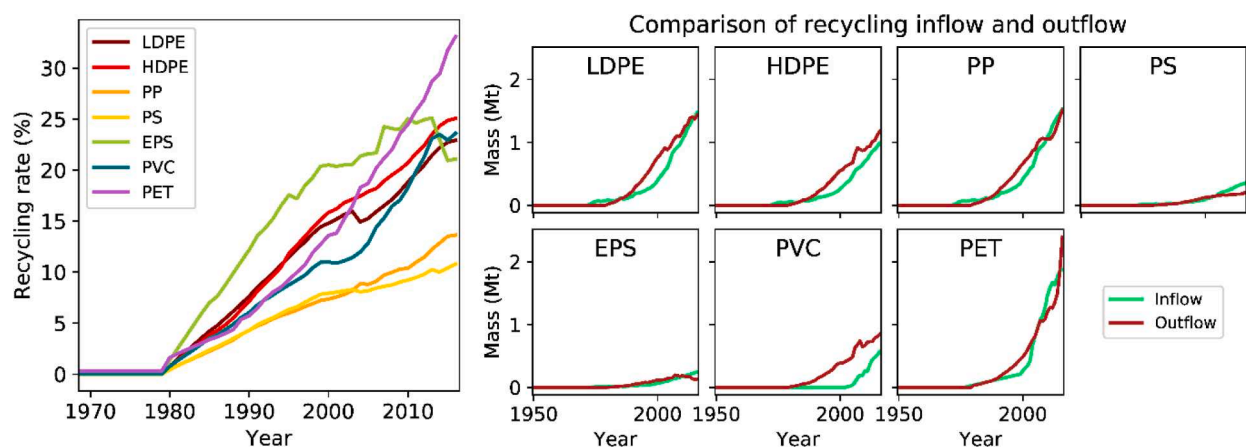


Fig. 9. Time series of flows of the seven polymers to recycling from 1950 to 2016. Graphs show the means of the probability distributions. Left: Recycling rates per material. Right: Comparisons of recycling inflows and outflows into and out of the system, as determined using two different modeling approaches.

the estimated uncertainty as the spread. The associated spread was estimated using a semi-quantitative method (Kawecki et al., 2018) depending on the type of literature, the material and the geographical entity it was valid for, the time difference, and whether it was

representative of the same process. The relative uncertainty could be calculated from the resulting probability distributions for flows when combining the mean and the standard deviation. The relative uncertainties obtained for the total inflows per compartment are displayed

separately for each polymer in Figures S1 to S7. The lack of data for years earlier than 1990–2000 is apparent in the larger relative uncertainty visible for all the polymers: proxy data used was treated with less confidence. Inflows into compartments after consumption stocks generally have lower relative uncertainties than inflows into compartments before consumption stocks as lifetimes influence the dynamics drastically. Combined with the higher quality of recent data, confidence in the results since 2000 remains high, despite the larger uncertainties of older data. In the last modeling decade, large uncertainties around 50%–60% were found for waste management flows for automotive applications for all polymers since the data used was not specific for plastics. The most uncertain flows for PET, PP, and HDPE (around 60% relative uncertainty) are for technical textiles, for which little material-specific data could be obtained. Flows out of the “Other” product sector were also associated with a higher uncertainty (50%–60%) as these included lesser-known and less used applications for which few precise data were available.

4. Discussion

The present work provides the first fully dynamic probabilistic material flow analysis (DPMFA) of seven important polymers used in Europe between 1950 and 2016, enabling the first calculations of the masses of polymer-specific stocks and sinks. To calculate these stocks, the lifetimes of specific product categories needed to be considered, not only their general product sectors, such as agriculture, as many different products with different lifetimes may have been used. The various polymers' contributions to product categories were very different, and thus simply using “plastics” as an input would not yield results as accurate as when using a polymer-specific approach. This approach may also have some value when looking at the global situation, as did Geyer et al. (2017). Although they used a dynamic model, those authors only considered eight very general product sectors to predict worldwide stocks and flows of “plastic”. However, as our model's results show, there were large differences between the uses, lifetimes, and end-of-life (EoL)-treatments of the different polymers.

The seven polymers considered in our study constituted 73.2% of the total European demand for polymers (excluding fibers) in 2016 (PlasticsEurope, 2018). Therefore, even though the analysis was incomplete, it included the major polymers in use today and covered all their major uses in different product sectors. The list of polymers contained some with a rapid turnover, such as packaging, and some with long lifetimes, such as those used in building, electronics, and the automotive sector. The flows for each of the plastics in our model were divided into nine product sectors covering 35 product categories and ten waste collection systems. Hence, this model provided a much higher resolution than previous MFA systems that mostly stopped at the product sector level, such as packaging or electronics. For example, Ciacci et al. (2017) only used six product sectors in their model of European PVC stocks and flows, and Jiang et al. (2020) considered the same number in their analysis of five polymers in China (PE, PP, PVC, PS, and ABS). However, there are many different types of packaging with very different EoL fates. Whereas PET bottles have a very high recycling rate, other plastic packaging mainly ends up in household waste. Moreover, different polymer uses in construction can have very different lifetimes: windows are more often replaced than pipes embedded into concrete structures. In agriculture, mulch films have a very short lifetime, whereas drainage pipes can have a life expectancy of many decades. A more detailed categorization of plastic's uses should, therefore, result in a more accurate description of polymer stocks. The approach developed in the present work should also be extended to other polymers with high production volumes that we did not cover, such as polyurethane or polyamide.

Including synthetic textiles in the MFA helped create a comprehensive base from which to develop a plastic emissions inventory. Since textiles have been found to be a major source of microplastics in the

environment, due to wear, washing, and drying (Cai et al., 2020; Carney Almroth et al., 2018), textile and non-textile applications need to be analyzed together. Dynamic MFA may help predict the total amount of plastic and microplastic released into the environment so far and thus model the amount stored in sinks such as soils or sediments. The study results revealed that using overall recycling, landfilling, or incineration rates failed to capture the true picture as there were marked differences between the polymers. For example, European recycling rates vary from 11% for PS to 33% for PET: using a single average value cannot capture the particularities of the recycling system. According to Eurostat, 42% of plastic packaging waste is recycled, but packaging only constitutes 39.7% of all plastics used (PlasticsEurope, 2018). PlasticsEurope reported an overall plastic recycling rate of 31.1% in 2016 (PlasticsEurope, 2018)—much higher than what is reported in our study, in which only PET comes close to this rate. One reason for this discrepancy may be the definition of recycling. Is it just the separate collection of waste—the flow into “Waste collection systems” in our model—or is it the actual flow into recycling systems or reuse? The recycling rates given by PlasticsEurope do not specify what recycling actually means. Haupt et al. (2017) have discussed the differences between collection and recycling rates in detail. They stated that for most of the materials collected separately and investigated in their study, the actual recycling rates determined using a systematic MFA were substantially lower than those officially communicated, which were often based on collection rates. Given the large discrepancy between the plastic recycling rates in our MFA and those officially reported, it is likely that the latter's use of “recycling” refers to separately collected materials. A certain fraction of separately collected waste is sent to incineration or landfill and is not counted as recycled in our model. Our model's mean polymer collection rate (including textiles) was 23%, ranging from 12% of PP and PS to 28% of PET and 58% of EPS. The low PET collection rate may seem surprising given the high PET recycling rates typically reported up to 60% (Welle, 2018), but this rate only refers to PET bottle recycling. PET, however, is used in many other applications, such as in the “Other Consumer Packaging” category, from which only 3% is collected separately. Our rate for PET also included all the textile uses that were completely ignored in most other plastic recycling studies.

The study's probabilistic approach enables both parameter uncertainty and parameter variability to be incorporated into the model. For example, in Table 1, we provided the standard deviations extracted from the probability distributions for the major polymer stocks and sinks. Previous dynamic MFA studies for polymers included no assessment of the range of possible results. Studies of PVC (Ciacci et al., 2017) and various polymers in China (Jiang et al., 2020) only gave single numbers for flows or stocks. Jiang et al. (2020) performed a sensitivity analysis by changing the parameters by $\pm 10\%$, but this only helped identify critical parameters and did not reveal the uncertainty associated with each result. Our results showed that the greatest part of the uncertainty originated from parameters treated as more uncertain using the Pedigree matrix because of limited data availability. Examples include EoL vehicle recycling practices, which were based on an incomplete data set from Eurostat, or the textile product sector for PET, which was based on global data. Also, our construction categories exhibited greater relative uncertainty as only old data or information for a different geographical unit were available. However, the identification of the most uncertain parameters paves the way for making future improvements of the model to target them.

Data from different regions of the world or even worldwide averages are difficult to compare with our results. For example, Geyer et al. (2017) used a quite simple dynamic MFA to calculate the fate of all the plastic ever produced in the world. According to their model, 30% of all the plastic ever produced in the world was still contained in the in-use stock, 12% had been incinerated, 10% had been recycled, and 60% had been landfilled or released to the environment. Our model's corresponding values for Europe were 19% in the in-use stock, 16% incinerated, 11% recycled, and 47% landfilled. However, as the average

world values combine very different regions—some with demanding recycling standards and some with only rudimentary waste collection infrastructure—comparison with our European values is difficult. Besides, it is likely that the very general approach used by Geyer et al. (2017), using officially reported “recycling rates” may overestimate actual recycling, as discussed above. Jiang et al. (2020) assessed the fate of five polymers in China using a dynamic MFA with broad product sectors. They reported limited polymer-specific data for PE, PP, PVC, PS, and ABS. The total in-use stock for those five polymers was 219 kg/cap, with 60% contained in buildings and constructions. Stocks in 2017 were estimated to be 74 kg/cap of PVC, 54 kg/cap of PE, and 50 kg/cap of PP. The corresponding values for our European model were 163 kg/cap of PVC, 128 kg/cap of PP (LDPE and HDPE combined), and 81 kg/cap of PP. Given Europe’s higher GDP than China and its longer accumulation of stock, our larger European polymer stocks are understandable. Giacci et al. (2017) previously determined Europe’s 2012 PVC stock to be 270 kg/cap, but their model only considered six very general product sectors and may thus have incorporated greater uncertainty than our more refined model with more specific product categories.

Our model included no release to the environment other than a very small release to wastewater from down-the-drain personal care products, for which release into wastewater is part of intended use. Other release processes, such as the polymer fibers shed from textiles during washing, were not included as this is an unintended consequence of washing. However, the previous static MFA, on which the present dynamic assessment was based, was the foundation of an assessment of plastic and microplastic release into Switzerland’s environment (Kawecki and Nowack, 2019). The dynamic mass flow analysis for Europe reported in this paper can now form the basis of an assessment of the release of plastic and microplastic over time in the European region. Such an analysis requires very detailed information on the life-cycles of specific products—not just product sectors—as release is very dependent on product type and use. For example, only very specific types of packaging are consumed on-the-go and can therefore be littered (Kawecki and Nowack, 2019); packaging that is used indoors will end up in household waste.

The drive towards a circular economy for plastics demands that policymakers and regulators have access to models that can identify critical product categories and waste streams when predicting the effects of potential measures. The regulation of plastic pollution has shifted in the last years from early bans of specific product categories such as plastic bags and regulation of waste handling, to a transition toward a circular economy (Syberg et al., 2021). For interventions at the level of specific product categories, a full life-cycle model is needed in order to identify those parts of the system that promise the largest possible effect. Our MFA shows that it is possible to track the flows of specific polymers resolved into detailed product categories. A complete model such as the one presented in this publication can form the basis onto which future interventions can be based, as it allows to predict not only the flows but also incorporates a detailed quantification of the consumption stocks. This is of special importance for all interventions targeting recycling. The much lower actual recycling rates resulting from our model question some of the targets given for recycling and put into legislation (Syberg et al., 2021) and call for a harmonization of the reporting of recycling rates and their clearer and unambiguous definition. Most of the waste generated per year comes from in-use stocks. Detailed knowledge on the magnitude and composition of the stocks, need to be taken into account to predict the effect of changes in policy when handling specific polymers or products. With the “European Strategy for Plastics in a Circular Economy” that has been launched by the EU in 2018 (European Commission, 2018), plastics recycling has become a key action point in the transition to a more circular economy. It has been shown that within the concept of circular economy, knowledge on the material stocks and the use phase is largely missing (Harris et al., 2021). The detailed assessment of historic and current stocks and flows of seven important polymers provided in this work can therefore also provide a solid basis to

move towards a more circular plastics economy.

5. Conclusions

The use of a dynamic probabilistic material flow analysis method enabled us to model the flows of seven different polymers through the European anthroposphere. Their stocks and sinks were quantified across nine product sectors and 35 product categories, providing a much higher resolution of the type of product categories than previous studies considering just a few very broad categories such as “packaging” or “construction”. This higher resolution, using distinct product categories with different life-cycles, enabled a more detailed analysis of polymer stocks and flows than previous assessments. For example, in our model, the “packaging” category was divided into seven sub-categories, each with very different uses and EoL treatments. For an accurate prediction of the effects of specific policy interventions about plastics use or for EoL options aiming to achieve a circular economy, the mass flows of specific products need to be understood in as much detail as possible. The dynamic MFA method for plastic flows present here delivers this information transparently, without losing sight of the bigger picture in the overview. Also, the environmental release of plastics can only be assessed using a model based on highly resolved product categories because interventions must target very specific product categories.

This detailed analysis of polymer life-cycles, stocks, and flows also needs to drill down to the level of specific polymers, as different polymers have very different uses and product distributions. Specific product categories and their very different EoL treatments and lifetimes are major determinants of polymer stocks and flows. For example, landfill is the major EoL compartment for LDPE, HDPE, PP, PS, and PET (48%–60%), whereas the largest fraction of EPS and PVC is still contained in in-use stocks (51% and 39%, respectively). These large differences clearly call for more polymer-specific assessments and moving away from just referring to “plastic” when discussing options to improve the circularity of various polymers.

Model availability

The Python scripts and SQL database used to perform the calculations are available at <https://github.com/empa-tsl/plastic-dpmfa>

The full results of the DPMFA are available at <https://doi.org/10.5281/zenodo.4756698>

Author contributions

The initial data was gathered by Qie Wu, who also performed the uncertainty assessment on the input data and the preliminary data analysis. Delphine Kawecki combined the datasets into a common, transparent model, set up the database, performed the final data analysis, and contributed to writing the article. João Gonçalves supported the development of the new methods used in the database and DPMFA package. Bernd Nowack wrote most of the article and supervised the project’s implementation and development.

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Delphine Kawecki: Methodology, Data curation, Validation, Software, Writing- Reviewing and Editing

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at [doi:10.1016/j.resconrec.2021.105733](https://doi.org/10.1016/j.resconrec.2021.105733).

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